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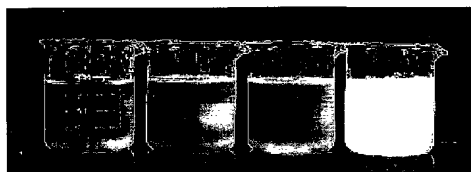
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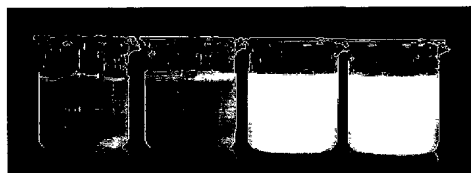
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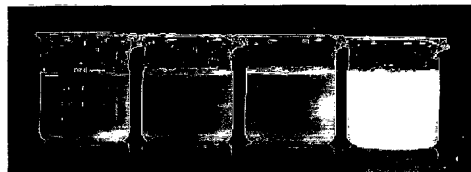
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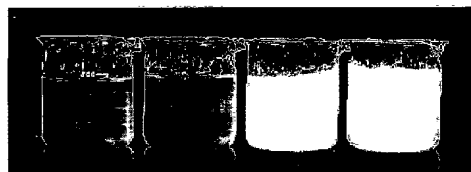
PHOSPHOROUS
ACID



PHOSPHORIC
ACID



ACETIC ACID



INORGANIC
SALT ACID

(57) Abstract: The present invention relates to a stained release composition of a biologically active substance, and a preparation method thereof. The sustained release composition includes an absorbing carrier having 0.0005~unit by weight of the biologically active substance adsorbed in 1000 unit by weight of a porous carrier, and a sustained release layer having a mixture of 0.05~unit by weight of polysaccharide, 0.3~unit by weight of inorganic alkali, and 0.25~unit by weight of one or more release regulators made of either organic or inorganic acid selected from the group consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, the mixture coated on the surface of the adsorbing carrier.

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A SUSTAINED-RELEASING AGRICULTURAL CHEMICAL AND THE METHOD FOR PRODUCING THEREOF

5 Technical Field

The present invention relates to a sustained release composition containing biologically active substance and a preparation method thereof. More particularly, the invention relates to a biologically active sustained release composition having biologically active substances of
10 agricultural chemicals or fertilizers, a coating matrix and a release regulator contained in a porous carrier, and a preparation method thereof.

Use of the composition according to the present invention can control a manifesting time of effects of agricultural chemicals or fertilizers, and can provide agricultural chemicals or fertilizers with reduced harmful
15 damages.

Background Art

Agricultural chemicals used to prevent damages by blight and harmful insects in vegetables, fruits, flowers or crops and fertilizers used
20 as an important source of nutrient for these crops are generally in a fluid form mixed with water, or granular form mixed with a filler. In such cases, agricultural chemical or fertilizer components may diffuse out of a sprinkled area, may retard in dispersion or may be evaporated, so that the concentration of effective components rapidly decreases. Since the
25 normal duration of efficacy is very short, fertilizers are generally sprinkled several times in a larger concentration than actually needed. Problems

with the use of excessive agricultural chemicals or fertilizers include various harmful damages to the health of crop producers or consumers, and serious harm to the environment due to salt accumulation or hypereutrophic state due to continued sprinkling or irrigation of fertilizers.

5 Accordingly, in order to maintain the efficacy of a biologically active substance of agricultural chemicals or fertilizers, in either solid or liquid form, for a long time, just by sprinkling once the biological active substance once in an appropriate concentration, providing a sustained release property thereto, research into methods for controlling an activity
10 manifesting time of biologically active substances is actively being carried out.

Known methods of preparing sustained release agricultural chemicals include: 1) encapsulating an agricultural chemical active substance into a microcapsule; 2) entrapping active substance of an
15 agricultural chemical into cyclodextrin; and 3) coating a resin on particles produced by mixing particulate or powdery active substances of agricultural chemical compositions alone or in combination with an extender.

Japanese Patent No. hei 6-116103 discloses a method of
20 providing a sustained release property by introducing a biodegradable resin agricultural chemicals dissolved in a solvent to a plate-injected biodegradable resin injected in a plate shape. Japanese Patent No. hei 5-85902 describes a method of preparing sustained release agricultural chemicals by mixing a raw material of an agricultural chemical with a
25 biodegradable polymer, dissolving the mixture in chloroform, adsorbing the resultant product into particulate zeolite, and heating to evaporate

chloroform therefrom.

U.S. Patent No. 4,647,537 discloses a method of encapsulating plant pathogenic microorganisms in a carrageenan polymer matrix. U.S. Patent No. 4,382,813 describes coagulation or precipitation of entrapped
5 insecticide by rapidly insolubilizing starch alkoxide containing divalent cations selected from the group consisting of calcium, barium, and strontium.

Korean Patent Publication No. 1989-1145 describes coating of particulate agricultural chemicals, wherein the particulate agricultural
10 chemicals are primarily coated with a mixture of isocyanate and flowable paraffin and then secondarily coated with organic or inorganic powder.

In addition, there are several known techniques of sustained releasing compounds.

Korean Patent Publication No. 1989-4995 describes that
15 agricultural chemical active substances are coated with solid, water-insoluble, low-melting point soldering materials and immobilized in a particulate fertilizer.

Korean Patent Publication No. 1992-7002910 describes a method of forming a polyurea barrier layer by performing interface
20 polymerization between a carrier containing agricultural chemicals and a polyhydroxylated compound or polyisocyanate.

Korean Patent Publication No. 2000-42895 describes mixing agricultural chemical active substance with a biodegradable resin and a release controlling agent and injection-molding the same.

25 Korean Patent Publication No. 2000-2248 a sustained releasing agent of increasing the sustained release property of fertilizers or

agricultural chemicals, by which the sustained release property of active substances immobilized in a matrix, e.g., pulp including waste paper, by immersing the active substances of fertilizers or agricultural chemicals into the matrix and forming a first resin coating and a second sulfur
5 coating.

However, the above-referenced methods have following problems. First, preparation processes are complicated and their applications are limited. Second, manufacturing cost is too high to be used for improvement of agricultural chemicals or fertilizers. Third, use
10 of solvents in preparing these materials may cause harmful surroundings. Fourth, in actual practices, coatings may rapidly biodegradable, making it difficult for these materials to exert sustained release effects.

In other words, some agricultural chemical compounds may not be used as sustained release composition because there may be
15 agricultural chemical compounds incapable of encapsulating or forming a clathrate compound with cyclodextrin. Also, since it is not possible to obtain a satisfactorily sustained release property while employing a conventional preparation method of a sustained release composition. Even if an agricultural chemical composition with a sustained release
20 property, the sustained releasing effect of the obtained agricultural chemical is insufficient. Thus, retention or extension of biological effectiveness of sustained release agricultural chemicals or reduction in harmful damage thereof cannot be fully accomplished. Further, since preparation methods of sustained release agricultural chemicals are
25 complicated and raw materials used in preparation thereof are expensive, there are many problems to be solved from the viewpoints of technology

and cost.

Therefore, there is gradually increasing demand for development of new sustained release agricultural chemical or fertilizer compositions and/or preparation methods thereof.

5 Meanwhile, although not devised for providing a sustained release property of agricultural chemical components, there are known several methods of coating polysaccharide on microorganisms. For example, Korean Patent Application No. 2000-17801 describes a method of coating polysaccharide derived from microorganisms on the
10 microorganism to provide heat resistance and acid resistance. Such microorganism coating techniques are for the purpose of protecting microorganisms advantageously used for human body, e.g., lactic acid bacteria, etc., from gastric acid and various intestinal digestive enzymes in the case of intake of the microorganisms microorganism, thereby
15 allowing the microorganisms to be safely seated in small and large intestines. Therefore, according to the microorganism coating method, while maintaining acid resistance, heat resistance and resistance to digestive enzymes, coating should be removed immediately when the microorganism reaches predetermined intestinal parts, e.g., small or
20 large intestines so that the microorganism can attach thereto and grow therein. Thus, the microorganism coating is quite different from the preparation process of sustained release agricultural chemicals, in which active substances of the sustained release agricultural chemicals are slowly released.

25

Disclosure of the Invention

To solve the above problems of the prior art, the present invention provides a sustained release composition of biologically active substances, which can be simply prepared, and a preparation method thereof.

The present invention also provides a sustained release composition of biologically active substances, which is cheap in raw materials and easily commercially available, and a preparation method thereof.

Also, the present invention provides a biologically active substance composition which is environmentally friendly and has a good sustained release property, and a preparation method thereof.

In an aspect of the present invention, there is provided a sustained release composition of a biologically active substance, the sustained release composition having an active substance adsorbed into a carrier, comprising an adsorbing carrier having 0.0005~50 unit by weight of a biologically active substance adsorbed in 1000 unit by weight of a porous carrier; and a sustained release layer having a mixture of 0.05~15 unit by weight of polysaccharide, 0.3~20 unit by weight of inorganic alkali, and 0.25~20 unit by weight of one or more release regulators made of either organic or inorganic acid selected from the group consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, the mixture coated on the surface of the adsorbing carrier.

In the sustained release composition of a biologically active

substance according to the present invention, the biologically active substance may be adsorbed into the porous carrier as described above. Otherwise, the biologically active substance may exist on the sustained release layer.

5 Also, the present invention provides a sustained release composition of a biologically active substance, comprising 1000 unit by weight of a porous carrier, and a sustained release layer having a mixture of 0.0005~50 unit by weight of a biologically active substance, 0.05~15 unit by weight of polysaccharide, 0.3~20 unit by weight of
10 inorganic alkali, and 0.25~20 unit by weight of one or more release regulators made of either organic or inorganic acid selected from the group consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, the mixture coated on the surface of the porous carrier.

 In the present invention, the inorganic alkali may be KOH or
15 NaOH.

 The sustained release composition according to the present invention may be adsorbed into the porous carrier coated with the biologically active substance or may be evenly distributed in a coating layer, that is, the sustained release layer. The release regulator
20 regulates release of the biologically active substance and controls biodegradation of the matrix (polysaccharide) for a considerable period, thereby allowing the active substance to be slowly released out of the sustained release composition.

 The present invention has the following effects. First, release is
25 sustained by adsorption between a carrier itself and active substance. Second, polysaccharide coating layer may sustain release of an active

substance. Third, a release regulator made of either organic acid or inorganic acid, such as phosphorous acid, phosphoric acid, acetic acid or hydrochloric acid, is first released to produce fine holes and the biologically active substance is then released through the fine holes, thereby enhancing a sustained release property and increasing a working convenience in sprinkling agricultural chemicals. Fourth, the release regulator contained in the sustained release layer sustains natural degradation (by microorganisms) of polysaccharide, thereby the sustained release property in actual practice.

Here, as the adsorbing carrier for agriculturally active components, one or more mixtures of natural minerals which are commonly used as main components of a filler and an extender in preparing soil-improving agents and agricultural chemicals, that is, zeolite, pearlite, vermiculite, diatomite, ceramic, sand and activated carbon. In addition, any carrier that has a soil affinity can be used as the adsorbing carrier. The carrier may be used in natural forms without any treatment, or in processed forms in which raw material is subjected to heat treatment at 600°C or higher, for removing internal impurity and optimizing the carrier state.

The porous carrier absolutely reduces diffusibility of biologically active substance by adsorbing the same and widens a contact area for preventing the sustained release layer coated on the surface thereof from stripping.

The sustained release composition according to the present invention is generally sprinkled to the soil manually or using a sprinkler. To this end, the diameter of a particle is preferably in the range of 0.5~5

mm, more preferably 2 mm or more in view of workability and utilization efficiency when applied to the soil. According to uses, the particle size may be increased or decreased.

In the present invention, the polysaccharide is naturally
5 degradable, and usable examples thereof include pestan, levan, xanthan gum, pullulan, polysaccharide-7, cellulose, zooglan, gellan, curdlan or suitable mixtures thereof.

Examples of the biologically active substance include an insecticide, a herbicide, a plant growth regulator, a nematocide, a
10 fungicide, a biocide, a raticide, a fumigant, an animal and insect repellent, a biological insecticide, pheromone, a sex stimulant, a flavoring agent, a deodorant, a dietary supplement, drug and fertilizers. In the sustained release composition according to the present invention, the amount of a biologically active substance can be suitably selected according to the
15 kind and specific activity of the biologically active substance.

Examples of agricultural chemical components include, but not limited to, an insecticide active component, a biocide active component, a herbicide active component and a plant growth regulator active component.

20 Examples of the insecticid active component include acephate, isoxathion, imidacloprid, ethylthiodemeton, ethofenprox, cartap, carbosulfan, clofentezine, cyclopyrifas-methyl, fenbutatin-oxide, cycloprothrin, dimetylrinphos, dimethoate, silafluofen, diazinon, thiodicarb, thiocyclam, tebufenozide, nitenpyram, vamidothion, bifenthrin,
25 pyridaphenthion, pyridaben, pyrimiphos-methyl, fipronil, phenisobromolate, buprofezin, furathiocarb, propafos, bensultap,

benfuracarb, formothion, malathion, monocrotophos, BPMC, CVMC, DEP, EPN, MEP, MIPC, MPP, MTMC, NAC, PAP, PHC, PMP and XMC.

Examples of the biocide active component include phosphorous acid salt, acibenzolar-S-methyl, azoxystrobin, bitanol, isoprothiolane, isoprodion, iminoctadine triacetate, oxolinic acid, oxone-copper, kasugamycin, carpropamid, captan, diclomezine, thiabendazole, thifluzamide, tecloftalam, tricyclazole, validamycin, hydroxyisoxazole, pyroquilon, fenarimol, ferimzone, fthalide, blasticidin, polyoxin, methasulfocarb), metalaxl, metalaxl-M, metominostrobin, mepronil, ampiciline, CNA, IBP, DF-351, NNF-9425 and NNF-9850.

Examples of the herbicide active component and plant growth regulator active component include azimsulfuron, atrazine, ametryn, inabenfide, imazosulfuron, nuiconazole, esprocarb, etobenzanid, oxadiazon, cafenstrole, quizalofop-ethyl, quinclorac, cumylron, chlomethoxylin, cyclosulfamuron, dithiopyr, cinosulfuron, cyhalofop-butyl, simazine, dimetametryn, dimepiperate, cinmethylin, dymron, thenylchor, triapenthenol, naproanilide, paclobutrazol, bifenox, piperophos, pyrazoxyfen, pyrazosulfuron-ethyl, pyrazolate, pyributicarb, pyriminobac-methyl, butachlor, butamifos, pretilachlor, bromobutide, bensulfuron-methyl, benzofenap, bentazon, benthocarb, bentoxazone, benfuresate, mefenacet, molinate, JA, SA, BABA, BTH, ACN, CNP, 2,4-D, MCPB, MCPB ethyl, and a plant growth regulator.

In the present invention, the biologically active substances may be used alone or in combination of a plurality of active substances.

Brief Description of the Drawings

FIG. 1 shows gelation characteristics of polysaccharides depending on the amounts of organic acid and inorganic acid;

FIG. 2A shows the biodegradation delay effect of polysaccharide, caused by the organic acid;

FIG. 2B shows the biodegradation delay effect of polysaccharide in the soil, caused by the organic acid;

FIG. 3 shows visual models of a sustained release composition;

FIG. 4 shows an elution test result of a sustained release composition model;

FIG. 5 shows another elution test result of a sustained release composition model;

FIG. 6A is a graphical representation showing a sustained release of salicylic acid;

FIG. 6B is a graphical representation showing a sustained release of ampiciline;

FIG. 6C is a graphical representation showing a sustained release of metalaxyl; and

FIG. 7 visualizes pesticidal effects of sustained release compositions.

Best mode for carrying out the Invention

Preparation methods of a sustained release composition according to the present invention will now be described. Solvents used throughout the description may be suitably selected from water, organic solvent or mixed solvent according to the polarity and stability of active

components. Since the solvent and water are evaporated and removed in the preparation process, the amount of the same used is not important and can be arbitrarily determined in consideration of workability.

5 **(I) Preparation of sustained release composition having an active substance adsorbed into a carrier**

The sustained composition according to the present invention having an active substance adsorbed into a porous carrier is prepared as follows.

10 First, 0.0005~50 g of a predetermined biologically active substance is dissolved in a predetermined volume, e.g., 100 ml, of a solvent according to the specific activity and desired activity thereof, thereby acquiring an active substance solution (Solution acquiring step). 1000 g of the porous carrier and the mixture is immobilized in the
15 acquired active substance solution to then be homogenously mixed and dried at 25~150°C, thereby obtaining an active substance adsorbing carrier (Immersing and drying step).

The obtained adsorbing carrier is homogenously mixed with the coating solution, the resultant mixture is coated on the surface of the
20 adsorbing carrier and dried at 25~150°C and a sustained release layer is finally formed (Coating step), thereby completing preparation of a sustained release composition of a biologically active substance according to the present invention.

Here, the drying temperature may be appropriately determined
25 according to the heat resistance of the active substance. From the

viewpoints of storage and workability, the drying is preferably performed such that moisture contained in the active substance is 40% or less.

In the present invention, coating of polysaccharide may be performed once or several times according to the kind and characteristics
5 of the active substance and the desired extent of sustained release.

(II) Preparation of sustained release composition having active substance contained in sustained release layer

A sustained release composition according to another
10 embodiment of the present invention is prepared in the following manner. Differently from the above, the preparation method of the present embodiment is simplified by simultaneously performing adsorption of active substance and coating of polysaccharide.

First, 0.0005~50 g of the biologically active substance is
15 dissolved in a predetermined volume, e.g., 50 ml, of a solvent, thereby acquiring an active substance solution (Solution acquiring step).

Separately, 0.05~15 g of suitable polysaccharide, 0.3~13 g of inorganic alkali, e.g., KOH, and 0.25~10 g of one or more mixtures made of either organic or inorganic acid selected from the group
20 consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, are homogenously mixed and a suspend coating solution is prepared (Coating solution preparation step)

The acquired active substance solution is homogenously mixed with the coating solution, thereby obtaining a coating solution containing
25 the active substance (Active coating solution preparation step). Then,

the coating solution containing the active substance and 1000 g of a porous carrier are homogenously mixed, and dried at 25~150°C so as to form a sustained release layer on the carrier (Coating step). Thereby, the preparation of a sustained release composition of a biologically active substance according to the present invention is completed.

Here, the drying temperature may be appropriately determined according to the heat resistance of the active substance. From the viewpoints of storage and workability, the drying is preferably performed such that moisture contained in the active substance is 40% or less.

10 In the present invention, coating of polysaccharide may be performed once or several times according to the kind and characteristics of the active substance and the desired extent of sustained release.

In the sustained release composition according to the present invention, an active substance is adsorbed into a porous carrier in a high concentration and diffusion of the adsorbed active substance is noticeably reduced due to the carrier, compared to the case where the active substance exists singly in a high concentration, thereby primarily attaining a sustained lease property. In the sustained release agricultural chemicals according to the present invention, natural polysaccharide is coated on the surface of an adsorbent having an active substance adsorbed thereinto or polysaccharide containing effective active substances is coated on/adsorbed into an adsorbing carrier. That is, the active substance and the carrier are incorporated and coated components are slowly decomposed, thereby exhibiting a sustained release property in a double manner.

Also, the organic and inorganic acids added as coating

components, e.g., phosphorous acid, phosphoric acid, acetic acid, hydrochloric acid and the like, prevent polysaccharide from rapidly decomposing, thereby maintaining durability of a sustained release layer and promoting biological active substances to be diffused out through
5 fine holes produced while the organic and inorganic acids are diffused out.

The present invention will now be described in more detail with reference to various examples. These examples are provided just for explaining the present invention in more detail and the invention is not
10 limited thereto.

For convenience, phosphorous acid salt was used as an active substance, zeolite was used as a carrier, curdlan was used as a coating agent derived from microorganisms, and KOH was used as inorganic alkali. However, the agricultural component, carrier and coating agent
15 used in the examples of the present invention are selected by way of example, and a variety of the above-referenced exemplary materials are also can be used. It is obvious to one skilled in the art that two or more combinations of agricultural chemical components can also be used.

20 **Preliminary Test Example 1: Gelation test of polysaccharide**

2.5 g of curdlan was added to 200 ml water and 8.3 g of KOH was added thereto and dissolved. 2.5, 5.0 or 10.0g of phosphorous acid (H_3PO_3), 2.5, 5.0 or 10.0g of phosphoric acid (H_3PO_4), 2.5, 5.0 or 10.0g of acetic acid (CH_3COOH), or 2.5, 5.0 or 10.0g of hydrochloric acid (HCl)
25 was added to the dissolved product and well stirred, followed by measuring pH levels of the respective resultant products. Then, the

samples were gelled (FIG. 1). The respective pH levels and extents of gelation are shown in Table 1.

As shown in the table, in the case where the organic acids were added in amounts of 2.5 g and 10 g, respectively, the pH levels were 13.1~13.3 and 3.4~5.6. Meanwhile, in the case where the organic acids were added in an amount of 5 g, the pH level for phosphoric acid was 11.7, but there was a negligible change in pH level for phosphorous acid and acetic acid.

The extent of gelation was sensitive to a change in pH, caused by addition of organic acid, rather than to the amount of organic acid added. In other words, to facilitate gelation, the pH level of a curdlan solution is preferably 12 or less.

【Table 1】

Organic acid	Amount added (g)	PH	Extent of gelation
Phosphous acid	2.5	13.2	-
	5	13.0	-
	10	5.61	Good
Phosphoric acid	2.5	13.1	-
	5	11.7	Good
	10	3.45	Good
Acetic acid	2.5	13.3	-
	5	13.2	-
	10	5.53	Good
Inorganic, hydrochloric acid	2.5	12.5	-
	5	2.0	Good

	10	1.0	Good
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**Preliminary Test Example 2: Biodegradation preventing test
1 of polysaccharide by organic acid**

(1) To investigate whether addition of organic acid delays
5 biodegradation of polysaccharide, starch was used.

As the organic acid, phosphorous acid, phosphoric acid and
acetic acid were used. Treated groups with organic acid added and a
control group without organic acid added were allowed to stand at room
temperature for 18 days, and biodegradation extents thereof were
10 examined. The composition details of the test groups and test results
are shown in Table 2.

As shown in the table, only the control group was biodegraded,
confirming that the organic acid considerably delayed biodegradation of
polysaccharide.

15 **【Table 2】**

	Starch solution	Other additives	Extent of biodegradation
Control group	2.5 g starch in 100 ml water		Biodegraded
Treated group 1	2.5 g starch in 100 ml water	8.3 g KOH, 10 g H ₃ PO ₃	No
Treated group 2	2.5 g starch in 100 ml water	10 g H ₃ PO ₃	No

Treated group 3	2.5 g starch in 100 ml water	8.3 g KOH, 10 g H ₃ PO ₄	No
Treated group 4	2.5 g starch in 100 ml water	8.3 g KOH, 10 g CH ₃ COOH	No

(2) 2.5g of curdlan was added to 200 ml water and well stirred, followed by heating at a microwave for one minute for gelation (Control group). 2.5g of curdlan and 8.3 g of KOH were added to 200 ml water and dissolved, followed by adding 10 g of H₃PO₃ and heating at a microwave for one minute for gelation (Treated group). The control group and treated group were allowed to stand at room temperature for 14 days and biodegradation extents thereof were examined.

The results showed that the treated group did not exhibit biodegradation and the control group was considerably biodegraded, confirming that the organic acid (phosphorous acid) considerably delays biodegradation of polysaccharide (FIG. 2A).

(3) If the sustained release composition according to the present invention is not biodegradable, it may undesirably cause environmental. In this regard, under the natural condition, that is, the condition in which phosphorous acid salt of the sustained release composition is repeatedly eluted, a time elapsed for occurrence of biodegradation was measured.

For elution of phosphorous acid from the treated group prepared in (2), the treated group was soaked in 400 ml water with an interval of 3 days, taken out of water, and then allowed to stand at room temperature. After the elution test was carried out 10 times, the treated group was biodegraded.

Thus, it was confirmed that biodegradation of the sustained

release composition coated with polysaccharide with an appropriate amount of organic acid added, was delayed in a natural state for a considerably extended period so that the sustained release composition maintained its sustained release property for some time and eventually
 5 was biodegradable.

(4) A time elapsed for occurrence of biodegradation of the coating components of the sustained release composition according to the present invention was measured under the conditions of nature.

2.5 g of curdlan was added to 200 ml water and 8.3 g of KOH
 10 was added thereto and well stirred, followed by heating at a microwave for one minute for gelation (Control groups AC, BC and CC). 2.5g of curdlan and 8.3 g of KOH were added to 200 ml water and dissolved, followed by adding organic acids in amounts listed in Table 3, and heating at a microwave for one minute for gelation (Treated groups
 15 A1~C3).

【Table 3】

	H ₃ PO ₃ (g)		H ₃ PO ₄ (g)		CH ₃ COOH (g)
A1	1.0	A2	0.84	A3	0.78
B1	0.5	B2	0.42	B3	0.39
C1	0.33	C2	0.28	C3	0.26

Each 5 g of the control groups and treated groups was planted 3 cm deep into the soil and kept at a plant cultivating greenhouse
 20 maintained at 20~25°C. After 3 months and 9 months, it was checked whether the samples remained in the soil or not.

FIG. 2B photographically shows the amounts of the control group and treated group remaining after 3 months. As shown in FIG. 2B, the treated group with more than 0.5 g of phosphorous acid, more than 0.42 g of phosphoric acid or 0.39 g of acetic acid added, remained in the soil
5 in a considerable amount.

Although not shown, after 9 months, there were no treated groups remaining in the soil.

Thus, it can be understood that organic acids can considerably delay the period of biodegradation of polysaccharide.

10

Preliminary Test Example 3 : Model test of sustained release composition

To visually confirm that the sustained release composition according to the present invention holds a sustained release property,
15 model tests were carried out.

As a carrier model with a biologically active substance adsorbed, a composition comprising methyl violet, which is widely used as a colorant for agricultural chemicals, was used. The methyl violet composition has 0.5 g of methyl violet adsorbed into 1 Kg of sand.

20 First, 8.3 g of KOH was dissolved in 100 ml of an aqueous solution and 2.5, 5 and 10g of curdlan were added to the resultant solution (Treated groups 1, 2 and 3), followed by adding 10 g of phosphorous acid, thereby preparing coating compositions, respectively. To the respective coating compositions were added each 1 Kg of methyl
25 violet composition, and uniformly stirred, followed by hot-air drying, thereby producing sustained release composition models. Uncoated

methyl violet composition was used as a control group. Photographs of the respective compositions are shown in FIG. 3.

Each 100 mg of treated group compositions was placed in a test tube and 5 ml deionized distilled water (DDW) was added thereto, and the resultant product was kept one day and then recovered. This procedure was repeated over 16 days, and changes in the color of recovered water were observed, as shown in FIG. 4, in which the respective test tubes are arranged in the order of recovered dates. Referring to FIG. 4, the uncoated composition underwent vigorous elution of methyl violet, that is, a large amount of methyl violet was eluted, from the first day, and the elution was nearly finished after the ninth day. On the other hand, in the treated groups, only a small amount of methyl violet was eluted and weak elution was continued over time. In particular, as the amount of curdlan added increased, the duration of release was increased and the amount of methyl violet eluted was reduced.

After 16 days elapsed, the respective compositions were recovered, dried, and colors of the remaining compositions were observed (FIG. 5). From the colors of coated compositions, it was understood that sustained release was continued even after the 16th day. As the more curdlan added, the more methyl violet remained. In the case of the control group, decoloration was severe, suggesting that methyl violet was mostly eluted.

Preliminary Test Example 4 : Preparation of sustained release layer containing active substance and sustained release

test

8.3 g of KOH was added to 200ml of an aqueous solution and 2.5 g of curdlan was dissolved therein, followed by adding 10 g of sodium salicylate as a signal substance for inductive resistance of a plant and uniformly stirring. Subsequently, 10 g of phosphorous acid was added to the resultant product for gelation, and dried at a dry oven maintained at 80~100°C for 20 minutes, thereby preparing a sustained release layer containing sodium salicylate.

35 mg of the sustained release layer containing sodium salicylate was placed into a test tube, 5ml DDW was added thereto, and the resultant product was kept one day and then recovered. This procedure was repeated, and each 50 μ l of the recovered samples was collected and the content of sodium salicylate contained in the sample solution was measured using high performance liquid chromatography (HPLC) (FIG. 6A).

The test was carried out in the same manner as described above, except that 2 g of ampiciline as an antibiotic was used as a biologically active substance, instead of sodium salicylate (FIG. 6B).

Alternatively, 10 g of metalaxyl was used as a biologically active substance, instead of sodium salicylate. Except that metalaxyl was first dissolved in 50 ml methanol, and mixed with a coating solution obtained by adding 10 g of phosphoric acid to 150ml of an aqueous solution containing 8.3 g of KOH and stirring the resultant mixture (FIG. 6C).

As shown, approximately one fifth (1/5) of the amount of release of the biologically active substance on the second day was continuously eluted even after 8 days.

Example 1: Preparation of sustained release composition having active substance adsorbed into carrier

10 g of metalaxyl was dissolved in 100 ml methanol to prepare an active substance solution (Solution acquiring step).

5 1000 g of dry zeolite was immobilized in the acquired active substance solution to then be homogenously mixed and dried at 25~150°C, thereby obtaining an active substance adsorbing carrier (Immersing and drying step).

Separately, 2.5 g of curdlan was added to 200 ml of water and 8.3
10 g of KOH was added thereto, followed by adding 10 g of phosphoric acid (H_3PO_4) and sufficiently stirring, thereby preparing a coating solution (Coating solution preparation step)

Then, the prepared adsorbing carrier was homogenously mixed with the coating solution, the resultant mixture was coated on the surface
15 of the adsorbing carrier and dried at 100°C such that moisture contained in the active substance is 35% or less, and a sustained release layer was finally formed (Coating step), thereby completing preparation of a sustained release composition of a biologically active substance according to the present invention.

20

Example 2: Preparation of sustained release composition having active substance contained in sustained release layer

10 g of metalaxyl was dissolved in 50 ml methanol to prepare an active substance solution (Solution acquiring step).

25 Separately, 2.5 g of curdlan was added to 200 ml of water and 8.3 g of KOH was added thereto, followed by adding 10 g of phosphoric acid

(H_3PO_4) and sufficiently stirring, thereby preparing a suspended coating solution (Coating solution preparation step)

Then, the prepared active substance solution and the coating solution were homogenously mixed, thereby preparing a coating solution
 5 containing the active substance (Active substance solution preparation step). The active substance solution and 1000 g of dry zeolite were homogenously mixed, the resultant mixture was coated on the surface of a dry adsorbing carrier and dried at 100°C such that moisture is 35% or less, thereby preparing a sustained release layer on the adsorbing carrier
 10 (Coating step) , thereby completing preparation of a sustained release composition of a biologically active substance according to the present invention.

15 **Application Example: Greenhouse test of sustained release agricultural chemicals**

To investigate whether the sustained release compositions of a biologically active substance prepared in Examples 1 and 2 according to the present invention actually exerted a sustained release effect in a greenhouse, this study was carried out.

20 4 sets each consisting of 24 stalks of healthy pepper plants aged 16 days from seeding, were prepared, and each set was treated with active substances in amounts listed in Table 4.

【Table 4】

	Treated amount per plant	Amount of active substance treated per plant (mg)
--	--------------------------	---

		Metalaxyl	Organic acid
Sustained release group 1	1 g of composition according to Example 1 (based on dry product)	10 mg	10 mg
Sustained release group 2	1 g of composition according to Example 2 (based on dry product)	10 mg	10 mg
Non-sustained release group	66ml of mixed solution of 150ppm metalaxyl and phosphoric acid	10 mg	10 mg
Untreated group	None	0	0

After 4 weeks from treatment, 10ml of a suspended solution of *Phytophthora capsici* zoospore of phytophthora blight was inoculated to each pepper plant in a concentration of $3.3 \times 10^3 \text{cfu/ml}$, which is approximately 3 times normal concentration. After 7 days from inoculation, the number of attacked stalks of pepper plants, onset rate and pesticidal activity were investigated (Table 5), and photographs corresponding thereto are shown in FIG. 7. In FIG. 7, the photograph marked by "A" shows sustained release group 2, the photograph marked by "B" shows a non-sustained release group, and the photograph marked by "CONTROL" shows an untreated group, respectively.

In the sustained release group 1, the result was similar to that of sustained release group 2. However, the photograph corresponding to

sustained release group 1 was not shown in the drawing, which is because it has been damaged during experimentation.

【Table 5】

	Number of attacked stalks	Onset rate (%)	Pesticidal activity (%)
Sustained release group 1	3	12.5	85.7
Sustained release group 2	4	16.7	80.9
Non-sustained release group	16	66.6	23.9
Untreated group	21	87.5	-

5 As understood from Table 5 and FIG. 7, even if the concentrations of the active substances treated are the same at an initial stage, the sustained release group with treatment of the sustained release composition according to the present invention has much higher pesticidal activity than the non-sustained release group. Also, even if
10 preparation methods used are somewhat different from each other, the sustained release compositions prepared in Example 1 and Example 2, respectively, have substantially the same sustained release effect and pesticidal activity.

Industrial Applicability

Use of the sustained release composition according to the present invention allows effective control of the period and amount of release of active substances of agricultural chemicals or fertilizers, thereby effectuating the effects of the agricultural chemicals or fertilizers. Therefore, it is not necessary to repeatedly apply heavily concentrated agricultural chemicals or fertilizers while maintaining the same efficiency.

Therefore, according to the present invention, the amount of agricultural chemicals or fertilizers used and agricultural labor force can be greatly reduced and harmful damages can almost be eliminated. Also, the sustained released composition according to the present invention has advantageous effects in terms of environmental preservation.

What is claimed is:

1. A sustained release composition of a biologically active substance, comprising:

an adsorbing carrier having 0.0005~50 unit by weight of the
5 biologically active substance adsorbed in 1000 unit by weight of a porous carrier; and

a sustained release layer having a mixture of 0.05~15 unit by weight of polysaccharide, 0.3~20 unit by weight of inorganic alkali, and 0.25~20 unit by weight of one or more release regulators made of either
10 organic or inorganic acid selected from the group consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, the mixture coated on the surface of the adsorbing carrier.

2. A sustained release composition of a biologically active
15 substance, comprising:

1000 unit by weight of a porous carrier; and

a sustained release layer having a mixture of 0.0005~50 unit by weight of a biologically active substance, 0.05~15 unit by weight of polysaccharide, 0.3~20 unit by weight of inorganic alkali, and 0.25~20
20 unit by weight of one or more release regulators made of either organic or inorganic acid selected from the group consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, the mixture coated on the surface of the porous carrier.

25 3. The sustained release composition of claim 1 or 2, wherein

the biologically active substance includes an insecticide, a herbicide, a plant growth regulator, a nematocide, a fungicide, a biocide, a raticide, a fumigant, an animal and insect repellent, a biological insecticide, pheromone, a sex stimulant, a flavoring agent, a deodorant, a dietary
5 supplement, drug and fertilizers. In the sustained release composition according to the present invention, the amount of a biologically active substance can be suitably selected according to the kind and specific activity of the biologically active substance.

10 4. The sustained release composition of claim 3, wherein the biologically active substance is a mixture of one or more compounds selected from the group consisting of phosphorous acid salt, acephate, isoxathion, imidacloprid, ethylthiodemeton, ethofenprox, bitanol, cartap, carbosulfan, clofentezine, cyclopyrifas-methyl, fenbutatin-oxide,
15 cycloprothrin, dimethylrinphos, dimethoate, silafluofen, diazinon, thiodicarb, thiocyclam, tebufenozide, nitenpyram, vamidothion, bifenthrin, pyridaphenthion, pyridaben, pyrimiphos-methyl, fipronil, phenisobromolate, buprofezin, furathiocarb, propafos, bensultap, benfuracarb, formothion, malathion, monocrotophos, BPMC, CVMC, DEP,
20 EPN, MEP, MIPC, MPP, MTMC, NAC, PAP, PHC, PMP, XMC, acibenzolar)-S-methyl, azoxystrobin, isoprothiolane, isoprodion, iminoctadine triacetate, oxolinic acid, oxone-copper, kasugamycin, carpropamid, captan, diclomezine, thiabendazole, thifluzamide, tecloftalam, tricyclazole, validamycin, hydroxyisoxazole, pyroquilon,
25 fenarimol, ferimzone, fthalide, blasticidin, polyoxin, methasulfocarb, metalaxl, metalaxyl, metominostrobin, mepronil, ampiciline, CNA, IBP,

DF-351, NNF-9425, NNF-9850, azimsulfuron, atrazine, ametryn, inabenfide, imazosulfuron, nuiconazole, esprocarb, etobenzanid, oxadiazon, cafenstrole, quizalofop-ethyl, quinclorac, cumylron, chlomethoxynil, cyclosulfamuron, dithiopyr, cinosulfuron, cyhalofop-butyl, simazine, dimetametryn, dimepiperate, cinmethylin, dymron, thenylchor, triapenthenol, naproanilide, paclobutrazol, bifenox, piperophos, pyrazoxyfen, pyrazosulfuron-ethyl, pyrazolate, pyributicarb, pyriminobac-methyl, butachlor, butamifos, pretilachlor, bromobutide, bensulfuron-methyl, benzofenap, bentazon, benthocarb, pentoxazone, benfuresate, mefenacet, molinate, JA, SA, BABA, BTHACN, CNP, 2,4-D, MCPB and MCPB ethyl.

5. The sustained release composition of claim 1 or 2, wherein the porous carrier includes one or more release regulators selected from the group consisting of zeolite, pealite, vermiculite, diatomite, ceramic, sand and activated carbon.

6. The sustained release composition of claim 1 or 2, wherein the polysaccharide is a mixture of one or more compounds selected from the group consisting of pestan, levan, xanthan gum, pullulan, polysaccharide-7, cellulose, zooglan, gellan and curdlan.

7. A method of preparing a sustained release composition of claim 1, comprising:
acquiring an active substance solution by dissolving 0.0005~50 unit by weight of the biologically active substance in a predetermined

volume of a solvent;

homogenously mixing the active substance solution and 1000 unit by weight of the porous carrier, drying and acquiring an active substance adsorbing carrier;

5 mixing 0.05~15 unit by weight of polysaccharide, 0.3~20 unit by weight of inorganic alkali, and 0.25~20 unit by weight of one or more release regulators made of either organic or inorganic acid selected from the group consisting of phosphorous acid, phosphoric acid, acetic acid and hydrochloric acid, the mixture coated on the surface of the adsorbing
10 carrier in a predetermined volume of a solvent, and preparing a suspend coating solution; and

 homogenously mixing the active substance adsorbing carrier and the coating solution so as to coat the surface of the adsorbing carrier with the coating solution, drying the resultant product to form a sustained
15 release layer.

8. A method of preparing the sustained release composition of claim 2, comprising:

 acquiring an active substance solution by dissolving 0.0005~50
20 unit by weight of the biologically active substance in a predetermined volume of a solvent;

 mixing 0.05~15 unit by weight of polysaccharide, 0.3~20 unit by weight of inorganic alkali, and 0.25~20 unit by weight of one or more release regulators made of organic acid selected from the group
25 consisting of phosphorous acid, phosphoric acid and acetic acid, in a

predetermined volume of a solvent, and preparing a suspend coating solution; and

homogenously mixing the active substance solution with the coating solution and preparing a coating solution containing the active
5 substance; and

homogenously mixing the coating solution containing the active substance and 1000 unit by weight of the porous carrier, and drying the resultant product to form a sustained release layer on the porous carrier.

10 9. The method of claim 7 or 8, wherein the biologically active substance includes an insecticide, a herbicide, a plant growth regulator, a nematocide, a fungicide, a biocide, a raticide, a fumigant, an animal and insect repellent, a biological insecticide, pheromone, a sex stimulant, a flavoring agent, a deodorant, a dietary supplement, drug and fertilizers.

15

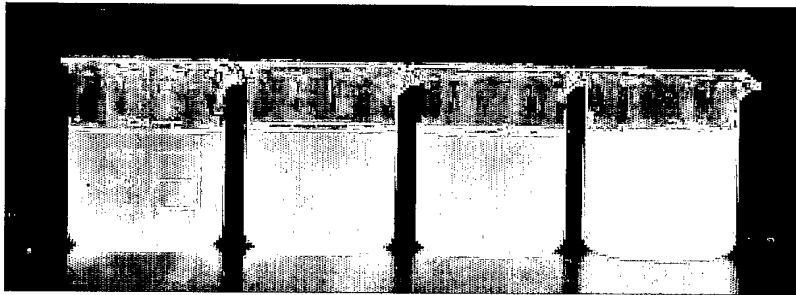
10. The method of claim 9, wherein the biologically active substance is a mixture of one or more compounds selected from the group consisting of phosphorous acid salt, acephate, isoxathion, imidacloprid, ethylthiodemeton, ethofenprox, bitanol, cartap, carbosulfan,
20 clofentezine, cyclopyrifas-methyl, fenbutatin-oxide, cycloprothrin, dimethylrinphos, dimethoate, silafluofen, diazinon, thiodicarb, thiocyclam, tebufenozide, nitenpyram, vamidothion, bifenthrin, pyridaphenthion, pyridaben, pyrimiphos-methyl, fipronil, phenisobromolate, buprofezin, furathiocarb, propafos, bensultap, benfuracarb, formothion, malathon,
25 monocrotophos, BPMC, CVMC, DEP, EPN, MEP, MIPC, MPP, MTMC, NAC, PAP, PHC, PMP, XMC, acibenzolar)-S-methyl, azoxystrobin,

isoprothiolane, isoprodion, iminoctadine triacetate, oxolinic acid, oxone-copper, kasugamycin, carpropamid, captan, diclomezine, thiabendazole, thifluzamide, tecloftalam, tricyclazole, validamycin, hydroxyisoxazole, pyroquilon, fenarimol, ferimzone, fthalide, blasticidin, polyoxin, methasulfocarb, metalaxl, metalaxyl, metominostrobin, mepronil, ampiciline, CNA, IBP, DF-351, NNF-9425, NNF-9850, azimsulfuron, atrazine, ametryn, inabenfide, imazosulfuron, nuiconazole, esprocarb, etobenzanid, oxadiazon, cafenstrole, quizalofop-ethyl, quinclorac, cumylron, chlomethoxynil, cyclosulfamuron, dithiopyr, cinosulfuron, cyhalofop-butyl, simazine, dimetametryn, dimepiperate, cinmethylin, dymron, thenylchor, triapenthenol, naproanilide, paclobutrazol, bifenox, piperophos, pyrazoxyfen, pyrazosulfuron-ethyl, pyrazolate, pyributicarb, pyriminobac-methyl, butachlor, butamifos, pretilachlor, bromobutide, bensulfuron-methyl, benzofenap, bentazon, benthocarb, pentoxazone, benfuresate, mefenacet, molinate, JA, SA, BABA, BTHACN, CNP, 2,4-D, MCPB and MCPB ethyl.

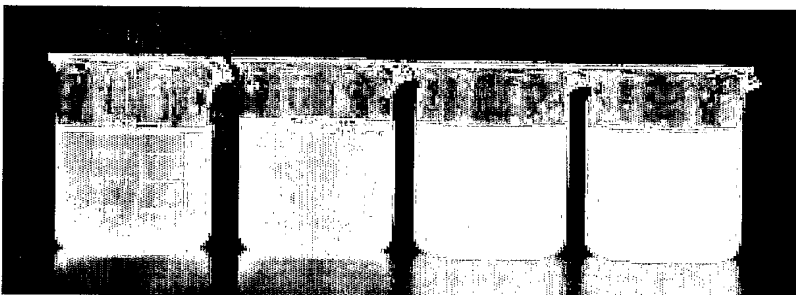
11. The method of claim 7 or 8, wherein the porous carrier is a mixture of one or more compounds selected from the group consisting of zeolite, pealite, vermiculite, diatomite, ceramic, sand and activated carbon.

12. The method of claim 7 or 8, wherein the polysaccharide is a mixture of one or more compounds selected from the group consisting of pestan, levan, xanthan gum, pullulan, polysaccharide-7, cellulose, zooglan, gellan and curdlan.

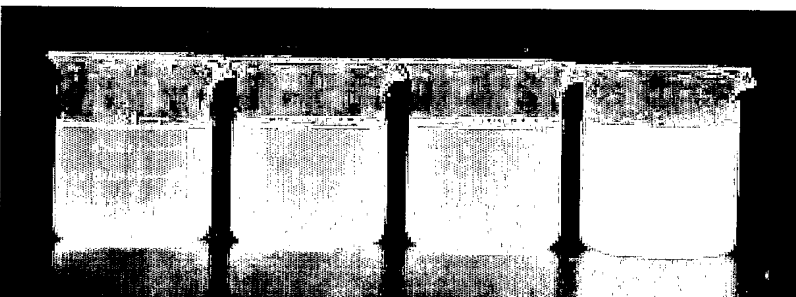
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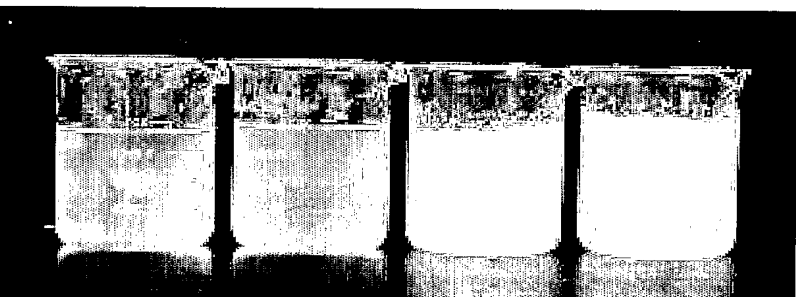
PHOSPHOROUS
ACID



PHOSPHORIC
ACID



ACETIC ACID

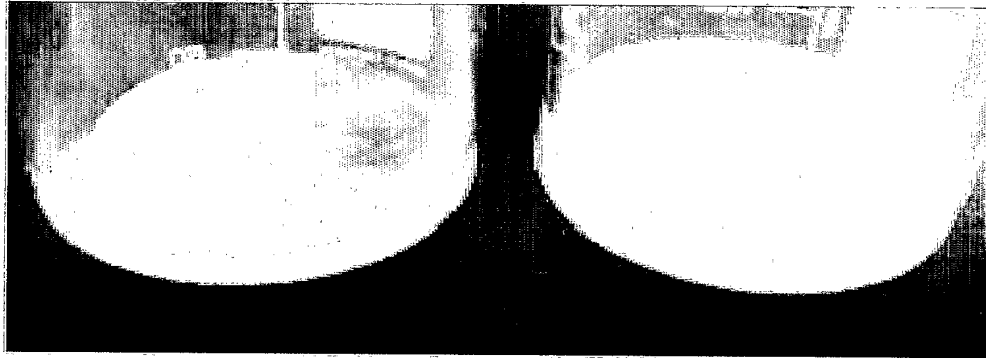


INORGANIC
SALT ACID

0g 2.5g 5g 10g

FIG. 1.

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CONTROL GROUP

TREATED GROUP

FIG. 2A.

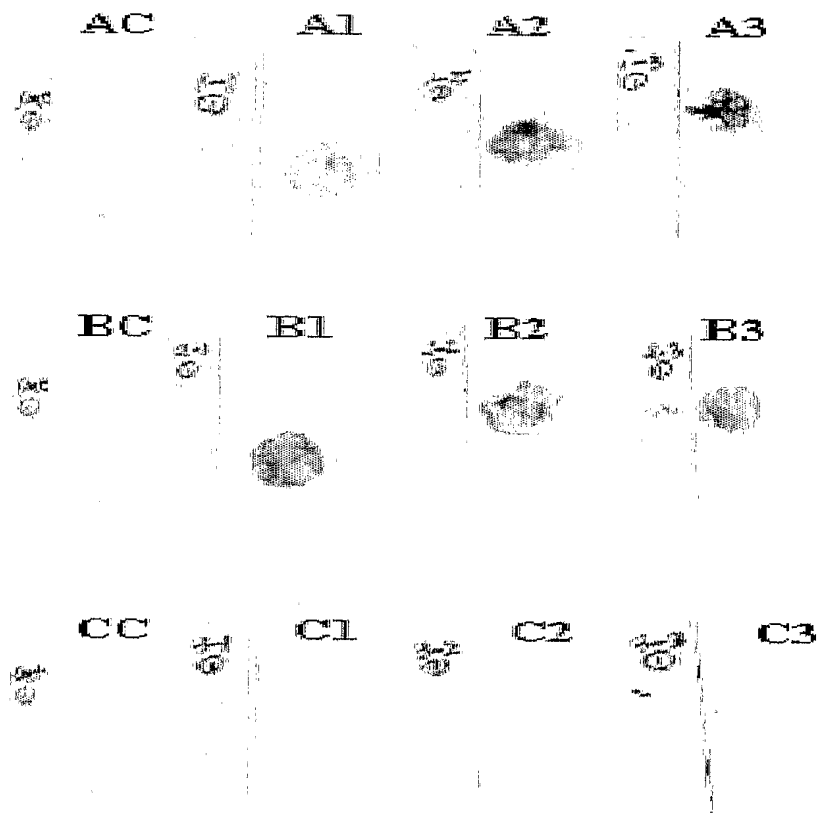


FIG. 2B.

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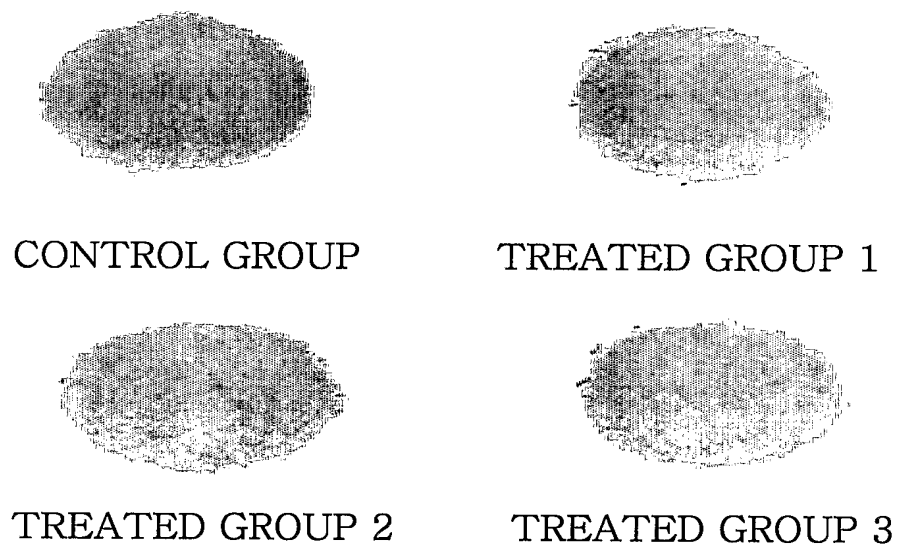


FIG. 3.

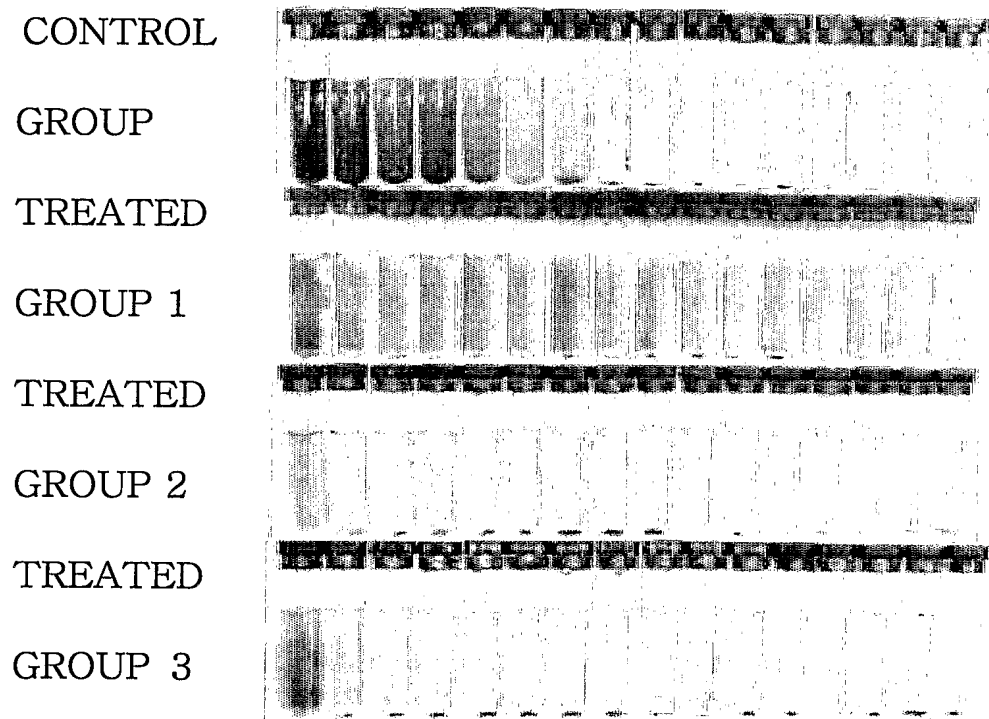


FIG. 4.

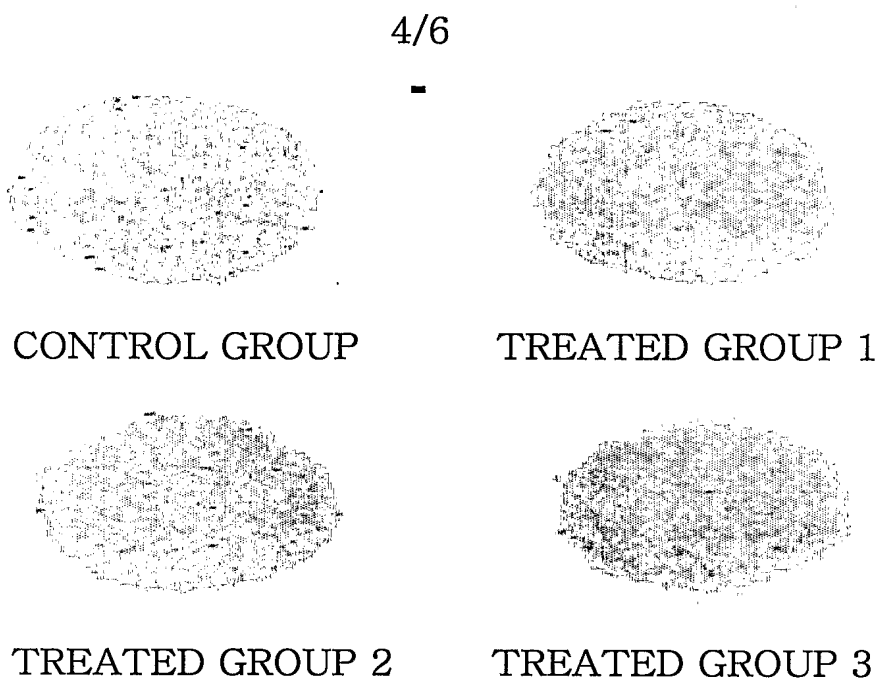


FIG. 5.

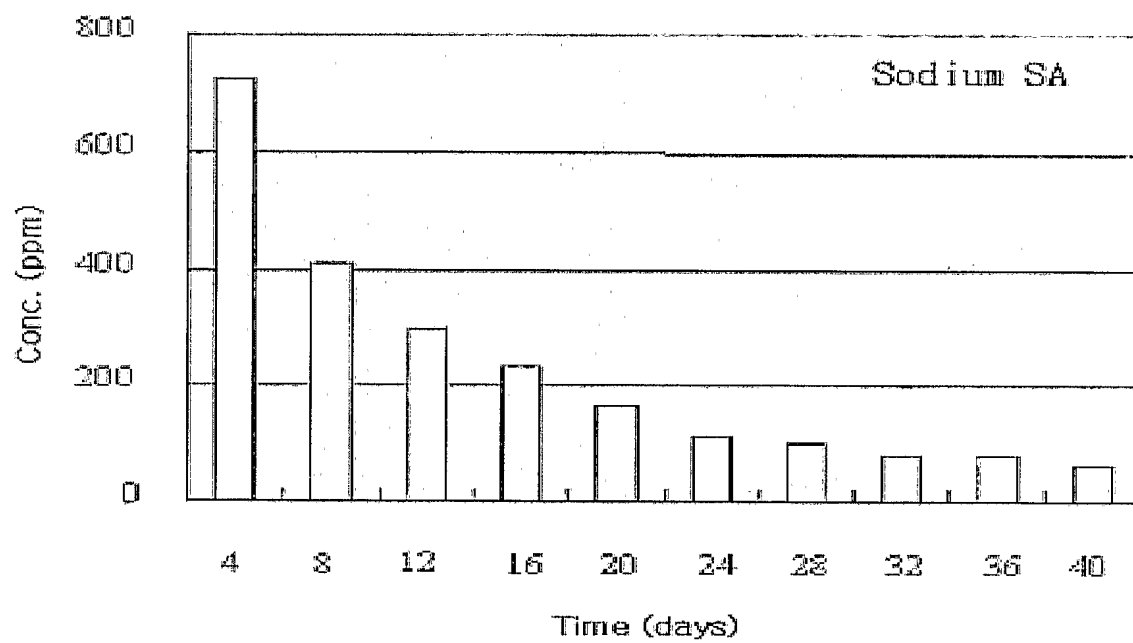


FIG. 6A.

5/6

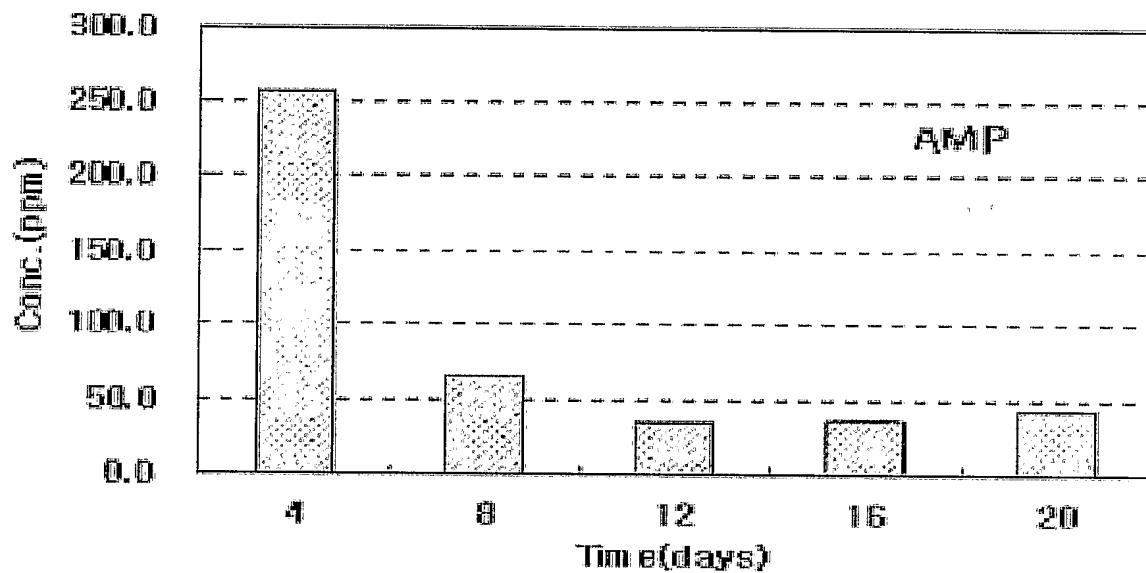


FIG. 6B.

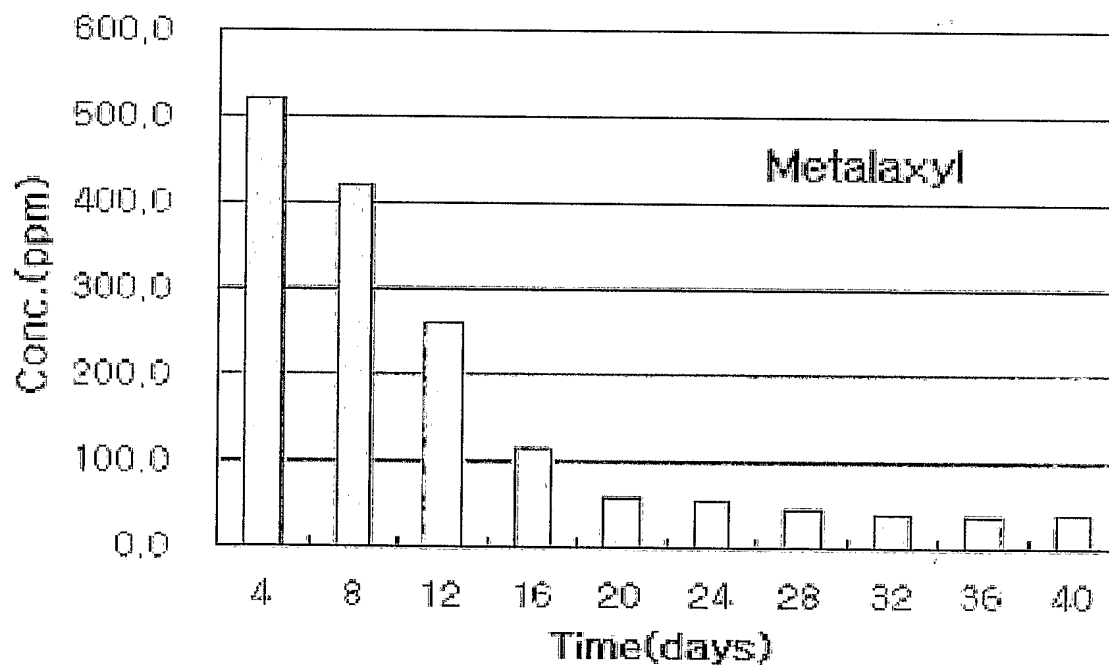


FIG. 6C.

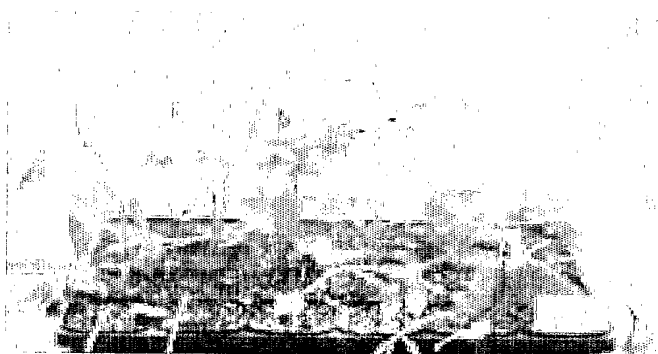
6/6



A



B



CONTROL

FIG. 7.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR03/00143

A. CLASSIFICATION OF SUBJECT MATTER

IPC7 A01N 25/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7 A01N 25/08, A01N 25/12, A01N 25/18, A01N 25/26

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean Patents and applications for inventions since 1975

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

NPS, PAJ, MEDLINE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	EP 364601 A (HOKKO CHEMICAL INDUSTRY CO., LTD) 25.04.1990 see the whole document	1-3, 5 4, 6-12
X A	WO 89/07889 A (HOKKO CHEMICAL INDUSTRY CO., LTD) 08.09.1989 see the whole document	1-3, 5 4, 6-12
Y A	JP 60-226801 A (KUMIAI CHEMICAL INDUSTRY CO., LTD) 02.12.1987 see the whole document	1-3, 5 4, 6-12
Y A	JP 56-169601 A (KUMIAI CHEMICAL INDUSTRY CO., LTD) 26.12.1981 see the whole document	1-3, 5 4, 6-12
Y A	JP 5-85902 A (JAPAN CHEMICAL INDUSTRY CO., LTD) 06.04.1993 see the whole document	1-3, 5 4, 6-12
A	KR 2001-0099583 A (PEPTRON CO., LTD) 09.11.2001 see the whole document	1-12
A	KR 2001-0022007 A (INFAIM THE TERA FJUTHICS) 15.03.2001 see the whole document	1-12

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

06 MAY 2003 (06.05.2003)

Date of mailing of the international search report

07 MAY 2003 (07.05.2003)

Name and mailing address of the ISA/KR



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Authorized officer

KWON, Oh Hee

Telephone No. 82-42-481-5597



INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR03/00143

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KR 2001-0022007 A	15.03.2001	NONE	